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TYPES AND COMPOSITIONS OF GLASS FOR PRODUCTION OF CONTINUOUS GLASS FIBER (REVIEW)

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Information on glass compositions currently used in the production of continuous glass fiber in Russia and abroad is supplied. The main physicochemical properties and areas of application of glass fibers are considered.

There are two known types of glass fiber: continuous and staple fibers, which differ in their properties, purpose, and production technology. Continuous fiber has an indefinitely large length and rectilinear parallel arrangement of filaments in strands, whereas staple fiber has a limited length and a tortuous and chaotic arrangement of filaments in space.

The process of continuous fiber formation from a melt via spinnerets is determined by the glass viscosity and its variation with temperature, the upper crystallization limit, and the rate of crystallization, as well as surface tension. It is expedient that the combination of these factors provide for a wide working interval and continuity of formation.

In the development of glass compositions, it is necessary to take into account the requirements imposed on the service parameters of the fibers, which are determined by the purpose and the production technology properties of the fiber. That is why glass is synthesized in various systems, which provides for qualitatively different properties in different fibers [1 – 3]. Table 1 indicates the compositions of the main types of glass fibers successfully used in industrial conditions, and Tables 2 and 3 indicate the main technological and service properties of glasses used to make glass fiber in Russia.

Glass A (neutral). More than 40 years ago, a substantial amount of fiber used to be made of soda-lime glass, which received the production name "composition A." Glass A is sometimes called "sheet glass," since it was nearly always produced by remelting sheet-glass waste. This composition has low chemical resistance to water and alkaline media and low strength.

Glass A cannot be used as a dielectric material. However, this glass fiber is cheaper than other types of fiberglass and can be used as a filler for fiberglass plastics when no stringent requirements are imposed. An example of glass A is neutral glass developed at the State Institute of Glass (GIS) and containing (wt.%): 71.0 SiO_2, 3.0 Al_2O_3, 8.5 CaO, 2.5 MgO, and 15.0 Na_2O [3].

Glass fiber of type A is not currently produced in Russia.

Glass E (electric-insulating). Alkali-free glass known as glass E and bearing not more than 1% R_2O is used in the production of continuous fiber to be used in manufacturing structural, insulating, and radio-engineering materials, which have a high thermal and moisture resistance. This glass was developed over 60 years ago by Owens Corning (U.S.) for electrical engineering. Later on, the area of application of this composition significantly expanded, and now nearly 90% of all fiberglass has the composition of glass E [1].

Glass E is calcium-aluminum-borosilicate and contains below 1% R_2O. The actual content of alkalis and the presence of traces of other elements, as a rule, depend on the choice of raw materials. Most glasses contain small quantities of fluorine, which facilitates the dissolving of raw materials, decreases the melt liquidus temperature, and improves fiber formation. The presence of ferric oxide in raw materials also has a significant effect on the stability of fiber formation, since iron ions increase the rate of infrared radiation of the melt, and the rate of heat transfer in glass exiting from the spinneret increases. All glasses used in the production of fiberglass of type E contain a relatively high (up to 10%) amount of boric anhydride, which is a costly and scarce component. Furthermore, the substantial volatilization of boric

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TABLE 1

Glass, patent number, company, country, published source	Weight content in glass, %									
	SiO_2	Al_2O_3	B_2O_3	CaO	MgO	TiO_2	ZrO_2	ZnO	$Na_2O + K_2O$	Fe_2O_3	F_2
Type A:											
Glass A (U.S.) [1]	71.8	1.0	–	8.8	3.8	–	–	–	14.2	0.5	–
Neutral, GIS (USSR) [3]	71.0	3.0	–	8.5	2.5	–	–	–	15.0	–	–
No. 65, VNIISV (USSR) [3]	60.0	3.0	–	8.0	3.0	–	6.0	6.0	12.0	2.0	–
No. 70, VNIISV (USSR) [3]	69.0	3.0	–	8.0	3.0	–	1.0	–	14.0	2.0	–
Type E:											
Standard alkali-free with 10% B_2O_3, VNIISV (USSR) [3]	54.0	14.5	10.0	16.5	4.0	–	–	–	0 – 1.0	0.5	0.3
Alkali-free with 8% B_2O_3, VNIISV (USSR) [3]	54.0	14.5	8.0	18.0	4.5	–	–	–	0 – 1.0	0.5	0.3
T-273A, VNIISPV (USSR) [4]	55.5	16.0	–	14.0	8.0	6.0	–	–	0 – 1.0	0.5	0.4
No. 2334961, Owens Corning Fiberglas (U.S.)	52.0 – 56.0	12.0 – 16.0	9.0 – 11.0	16.0 – 19.0	3.0 – 6.0	–	–	–	–	–	–
No. 621, No. 2571074, Owens Corning Fiberglas (U.S.)	52.0 – 56.0	12.0 – 16.0	8.0 – 13.0	19.0 – 25.0	–	–	–	–	–	–	Up to 3.0
No. 4542106, PPG Industries (U.S.)	58.0 – 60.0	11.0 – 13.0	–	21.0 – 23.0	1.0 – 4.0	1.0 – 5.0	–	–	0 – 1.0	–	–
No. 3037136, Nippon Sheet Glass (Japan)	54.5 – 57.0	13.0 – 16.0	5.0 – 7.5	21.0 – 23.0	0.6 – 3.0	0 – 1.0	–	–	0 – 1.0	0 – 1.0	–
ECRGLAS, Owens Corning Fiberglas (U.S.)	54.0 – 65.0	9.0 – 15.0	–	17.0 – 25.0	0 – 4.0	0 – 4.0	–	2.0 – 5.0	0 – 1.0	–	–
Advantex, No. 5789329, Owens Corning Fiberglas (U.S.)	59.9	13.5	–	22.3	3.2	0.2	–	–	0.3	0 – 1.0	–
Type C:											
No. 2308857, Owens Corning Fiberglas (U.S.)	65.0	3.8	5.5	13.7	2.4	–	–	–	8.5	0.3	–
No. 4628038, Owens Corning Fiberglas (U.S.)	53.3	16.0	3.0	15.8	2.5	0 – 2.0	–	–	7.0	0 – 2.0	–
No. 7, No. 289991, VNIISV (USSR)	64.0	5.5	–	12.0	2.0	2.0 BaO	2.0	1.7 Mn_3O_4	9.5	–	0.3
No. 7-A, No. 787382, VNIISPV (USSR)	64.0	4.5	–	12.0	3.5	0.2	4.2	–	11.5	–	0.3

anhydride in melting has a negative effect on the environmental safety of the production facility.

The standard domestic alkali-free aluminum-borosilicate glass E and similar glasses used abroad have a high chemical resistance to water (hydrolytic class I), good dielectric properties (specific resistance $10^{12}\ \Omega \cdot m$), sufficiently high strength (up to 3300 MPa), and a low TCLE (about $60 \times 10^{-7}\ K^{-1}$).

At present, various products are made from glass E: fiberglass fabrics and meshes for various purposes, non-woven materials and fiberglass plastics based on these materials.

However, glass E has a number of technological disadvantages. The most serious is the presence of two volatile components (boron oxide and fluorine), which intensely evaporate in melting. This often leads to the disturbance of the chemical homogeneity of the glass and, at the same time, pollutes the environment. The problem of volatilization of raw materials can be partly solved by electrical melting of glass under a cold batch layer. Another substantial shortcoming of glass E is the high aggressiveness of the glass melt with respect to the majority of refractories. The use of expensive refractories, as well as expensive and scarcely available boric acid, significantly increases the fiberglass production cost. Finally, the fiber made of glass E is insufficiently resistant to acids, which in some cases restricts its application.

Therefore, numerous researchers and fiberglass manufacturers have been developing low-boron and boron-free alkali-free glasses, which in their service properties are not inferior to glass E [6]. Some of the most successful developments in this field are the ECR and Advantex glass fibers developed by Owens Corning (U.S.) [7]. The viscosity curves of such glasses are 60 – 70°C higher than those of standard glass E. At the same time, Advantex glass surpasses the standard alkali-free aluminum-borosilicate glass in its resistance to chemical agents, and the strength of fiber made of this glass in corrosive media declines much more slowly than in glass E fibers [8, 9]. The Stekloplastik Company also has de-

TABLE 1 (continued)

Glass, patent number, company, country, published source	Weight content in glass, %										
	SiO_2	Al_2O_3	B_2O_3	CaO	MgO	TiO_2	ZrO_2	ZnO	$Na_2O + K_2O$	Fe_2O_3	F_2
Type D:											
D (U.S.) [1]	75.5	0.5	20.0	0.5	0.5	–	–	–	3.0	–	–
D-4.5, VNIISPV (USSR) [5]	51.0 – 71.0	1.0 – 5.0	25.0 – 45.0	–	–	–	–	–	3.0	–	–
No. 63002831, Nippon Electric Glass (Japan)	70.0 – 80.0	0 – 2.0	15.0 – 21.5	0 – 2.0	0 – 1.0	–	–	–	2.0 – 5.0	–	–
No. 8333137, Nitto-Boseki (Japan)	50.0 – 60.0	10.0 – 20.0	20.0 – 30.0	0 – 5.0	0 – 4.0	0.5 – 5.0	–	–	0 – 0.5	–	–
Type S:											
No. 3402055, Owens Corning Fiberglas (U.S.)	55.0 – 79.9	12.6 – 32.0	–	–	4.0 – 20.0	–	–	–	–	–	–
No. 3459568, PPG Industries (U.S.)	54.0 – 62.0	20.0 – 27.0	–	–	5.0 – 11.0	2.0 – 10.0	–	–	0 – 2.0 Li_2O	–	–
R, No. 1435073, Vetrotex (France)	55.0 – 65.0	20.0 – 30.0	–	5.0 – 20.0	2.0 – 10.0	–	–	–	–	–	–
No. 11021147, Nitto-Boseki (Japan)	60.0 – 70.0	17.0 – 27.0	–	–	7.0 – 17.0	–	–	–	–	0.1 – 0.5	–
VMP, VNIISPV (USSR)	58.0 – 73.0	15.0 – 25.0	–	–	4.0 – 15.0	0.3 – 2.8	0.3 – 0.7	–	–	0.5	–
VM-1, VNIISPV (USSR)	55.0 – 57.0	24.0 – 26.0	–	–	14.0 – 16.0	1.3 – 2.7	–	–	–	–	–
No. 2129102, Stekloplastik Co. (Russia)	57.0 – 60.0	20.0 – 27.0	–	–	10.0 – 16.0	0.2 – 0.7	0 – 0.2	–	0.1 – 0.4	0.1 – 0.6	–
Alkali-resistant glass:											
Cemfil, No. 1243972, Pilkington (Gr. Britain)	71.0	1.0	–	–	–	–	16.0	1.0 Li_2O	11.0	–	–
AR, No. 5307116, Kanebo Ltd. (Japan)	60.7	–	–	–	–	–	21.5	–	16.5	1.3 Li_2O	–
Shch-15Zh, Inventor's Certif. No. 451652, GIS (USSR)	65.8	5.6	–	7.4	–	–	7.4	–	9.0	4.7	–
Shch-15ZhT, Inventor's Certif. No. 874689, GIS (USSR)	63.0	4.1	–	9.2	0.3	6.2	3.5	–	8.3	4.7	–
No. 2083516, D. I. Mendeleev RKhTU (Russia)	50.0 – 60.0	12.0 – 23.0	–	5.0 – 14.0	0.5 – 11.0	–	0.1 – 0.5	–	12.5 – 19.0	–	–

veloped formulas for fiberglass production, namely, T-273A and Kt, which do not contain B_2O_3 and F_2 [4, 10].

Glass C (chemically resistant). The use of fibers made of glass E, which is the most abundant in the world, is unadvisable in certain types of fiberglass plastics, owing to its low resistance to acid and alkaline media and high cost and scarce supply of one of the batch components, namely, boric acid. Therefore, fiberglass of type C was developed for engineering areas, in which material is in contact with aggressive media, mostly, acids. There are many formulas for glass C, both in our country and abroad.

The main factors ensuring increased acid resistance and satisfactory water resistance in the glass are the following: at least 60% SiO_2 content; use of sodium and lithium oxides, whose ions have a high electric field intensity; the predominant use of calcium and zinc oxides as alkaline-earth oxides; introduction of small quantities of Al_2O_3, B_2O_3, Fe_2O_3, TiO_2, MnO, and SnO_2; the presence of oxides of multivalent ions insolvent in acids.

A fiber from alkaline glass satisfying these conditions used to be produced in the USSR (USSR Invertor's Certif. No. 289991). The fiber had good corrosion resistance in acids; however this glass had relatively inferior technological properties in molding glass beads and fiber (a high propensity to crystallization, a narrow molding temperature interval). Furthermore, the batch of this glass contained toxic components.

Composition 7-A (USSR Invertor's Certif. No. 787382) satisfied the above technological requirements best of all and was successfully used at Russian factories. With respect to its working properties, this glass corresponds to alkali-free glass E and ensures a stable fiber-forming process [11]. The fiber of this composition has high acid resistance and is more resistant to alkali than glass E. It is also important that glass 7-A has a low cost. At the same time, this fiber has low strength, which makes it unfit as a filler in high-strength fiberglass plastics. It cannot be used as an insulating material either.

TABLE 2

Glass	Weight content, %							
	SiO_2	Al_2O_3	B_2O_3	CaO	MgO	TiO_2	$Na_2O + K_2O$	F_2
E	52.0 – 55.0	14.0 – 15.0	7.0 – 10.0	17.0 – 22.0	2.5 – 4.0	–	0 – 1.0	0.3
Kt	58.0 – 60.0	12.0 – 14.0	–	20.0 – 22.0	3.0 – 5.0	1.0 – 2.0	0 – 1.0	–
7-A	63.0 – 65.0	4.0 – 5.0	–	11.0 – 13.0	3.0 – 4.0	4.0 – 5.0 ZrO_2	11.0 – 12.0	0.3
VM-1	55.0 – 57.0	24.0 – 26.0	–	–	14.0 – 16.0	1.3 – 2.7	–	–
VMP	58.0 – 73.0	15.0 – 25.0	–	–	4.0 – 15.0	0.3 – 2.8	–	–
Hollow	52.0 – 55.0	14.0 – 15.0	9.0 – 11.0	17.0 – 22.0	2.5 – 4.0	–	0 – 1.0	0.3
Quartz	99.95	–	–	–	–	–	–	–

Glass fibers of type C are used in the production of filtering materials, primarily, acid-resistant ones, and also in the production of roof mats and bitumen reinforcement. Glass C in the form of fibers and fabrics is used in composite materials that have contact with acid materials or serve as acid containers, for instance, galvanizing batch, storage battery tanks, etc.

Glass D (with low dielectric permittivity). The progress made in electronics and related industries generated a need for glass fibers with lower dielectric parameters than those of glass E. To solve this problem, glass D was developed. Most D glasses were synthesized on the basis of the $SiO_2 – B_2O_3 – Al_2O_3$ system (the B_2O_3 content can reach 30%) with up to 3% additive of alkaline metal oxides to facilitate glass formation, decrease the melting temperature, and increase the chemical resistance. The best technological and physical parameters are shown by glasses with an equal molar content of aluminum and sodium oxides [12]. Most often, the dielectric constant of such glasses is around 4.0 at room temperature. At the same time, type D fiberglass is characterized by a low level of strength and chemical resistance. Glass fiber of type D is used as a reinforcing material in electron boards and radar housing.

Glass S (high-strength, high-modulus). Reinforcing materials with high strength and elasticity modulus parameters are needed to develop composite materials with increased physicomechanical parameters, in particular, those of tensile and compressive strength. Since the elasticity modulus of binders is very low, the elastic properties of composites are determined by the type of glass fiber used as a filler.

There are several composite materials whose application areas require higher strength and hardness than fiberglass plastics reinforced with fiberglass of type E. Glass fiber of type S with high parameters of tensile strength and elasticity modulus have been developed especially for such composites. These fibers were developed on the basis of $MgO – Al_2O_3 – SiO_2$ and $MgO – CaO – Al_2O_3 – SiO_2$ systems, using BeO, TiO_2, and ZrO_2 in certain cases.

Experimental studies performed at the Steklopastik Company indicated that the synthesis of $MgO – Al_2O_3 – SiO_2$ glasses containing 57 – 73% silicon oxide makes it possible to produce fibers with a high level of strength (5900 – 7000 MPa) [13].

Glass fiber of type S exceeds the glass fiber of type E by about 20% in elasticity modulus and by nearly 40% in strength. Glass S has a softening point of 970°C, which makes it possible to operate materials based on this fiberglass at higher temperatures. Glass S has a lower dielectric permittivity than the insulating aluminum-boron-silicate glass. In corrosion resistance to acids, magnesium-aluminosilicate fibers exceed even the type-C fiberglass.

Typical features of high-modulus fibers in the specified region include a density of up to 2600 kg/m^3, an elasticity

TABLE 3

Parameter	Glass						
	E	Kt	7-A	VM-1	VMP	hollow	quartz
Density, kg/m^3	2570	2680	2610	2580	2560	1700	2210
Softening point, °C	840	900	780	920	945	840	1667
Strength, MPa	3445	3500	3350	4500	5000	2800	4000
Elasticity modulus, MPa	73,500	74,000	73,500	95,000	93,000	–	74,900
Weight loss in boiling, $mg/5000\ cm^2$:							
in H_2O	18	17	20	13	11	–	–
in 1 N HCl	1000	155	15	640	170	–	–
in 2 N NaOH	1250	310	550	1075	1040	–	–
Dielectric constant*	6.60	7.60	–	5.93	5.18	4.10	3.0 – 4.0
Loss tangent of dielectric*	39×10^{-4}	29×10^{-4}	–	–	–	100×10^{-4}	1.5×10^{-4}

* At a temperature of 20°C and a frequency of 10^6 Hz.

modulus of up to 117,000 MPa, and a strength of 3500 – 4500 MPa (glass VM-1) [14].

The production of high-strength and high-modulus glass fiber is a labor-consuming and costly process, since these glasses require high melting and working temperatures. Moreover, at higher temperatures, the service life of platinum materials used in drawing the glass fiber decreases. Therefore, the use of fiberglass of type S is limited. It is used mostly in the aircraft industry and in production of specialized sports equipment.

Alkali-resistant glass. It is known that concrete has low values of elasticity modulus, and metal reinforcement makes concrete too heavy. Therefore, an idea originated about 30 years ago of developing a composite material in which cement would be the matrix and fiberglass would be the filler. Integrated studies indicated that glass fiber significantly improves the structural and technological properties of concrete materials and improves the crack resistance and impact strength. However, production of glass concrete raises a serious problem of alkali resistance of the fiber, since the main component of the liquid phase of cement in hardening is a solution of calcium and sodium hydroxides.

Most formulas of alkali-resistant glasses for cement reinforcement lie within the $SiO_2 – ZrO_2 – Na_2O$ system with certain additives [15].

The Pilkington Brother Company (Great Britain) in the early 1970s developed and started industrial production of a high-zirconium fiberglass named Cemfil for cement reinforcement [16].

The GIS Institute has been carrying out systematic research in the synthesis of glasses suitable for cement reinforcement. As a result, glass Shch-15Zh (USSR Invertor's Certif. No. 451652) was developed virtually in the same system as Cemfil glass, containing iron oxides along with 10% zirconium oxide, as the authors believe that zirconium and iron oxides are the most effective additives increasing the fiber strength. Further research demonstrated that the alkali resistance of glass fiber increases with the ZrO_2 content in the glass composition increasing to 16 – 18% (USSR Invertor's Certif. No. 912703).

High-zirconium (up to 15 – 20% ZrO_2) glasses for continuous fiberglass production have certain disadvantages restricting their area of application. The high-melting high-zirconium compositions significantly complicate the fiber production technology. Furthermore, zirconium-bearing raw material is scarce and unacceptable for mass production. Accordingly, the Cemfil fiber is sold at a price two times higher than the price of standard glass fiber E. The mass-scale production of high-zirconium glass could be accomplished only in the case of using cheaper zircon-bearing waste generated by the mining industry [17].

The problem of refining glass formulas for cement reinforcement remains topical. Since cement-resistant fiber is intended for construction and has to be produced in large volumes at a relatively low cost, research is directed towards developing zirconium-free or low-zirconium glasses with good technological properties and sufficient resistance in cement [15]. The researchers at D. I. Mendeleev Russian Chemical Engineering University investigated alkali-resistant glasses with a low ZrO_2 content and without ZrO_2, in $Na_2O – MgO – Al_2O_3 – SiO_2$ and $Na_2O – MgO – Al_2O_3 – SiO_2 – TiO_2 – ZrO_2$ systems [18 – 20]. As a result, glass fibers resistant in cement medium were developed (USSR Invertor's Certif. Nos. 1511227 and 1512938; RF Patent No. 2083516).

Shielding glass fibers. The progress made in nuclear physics and space exploration generated the need for the development of glasses and glass fibers resistant to radiation effect. Since the capacity for absorbing radiation increases with increasing density of the material, the fibers used for this purpose contain components with a high atomic weight. Russian and foreign researchers developed this kind of glass and fiber with large quantities of PbO, CdO, CeO_2, BaO, Bi_2O_3, and B_2O_3. The strength of such glass fibers does not exceed 2500 MPa. They are characterized by high density and dielectric permittivity, but at the same time, have low strength and chemical resistance. The glass fibers resistant to radiation are used to make shielding fabrics and screens, as well as composite materials with shielding properties [14].

Hollow fibers. A technology for producing glass fibers with a modified geometry, i.e., with a hollow structure, was developed to produce lightweight fiberglass plastics and composites with improved radioengineering properties. Such fibers are made of glass E. An important parameter of hollow fiber is the capillary coefficient, which constitutes the ratio of the inner diameter of the fiber to its outer diameter, which is equal to 0.5 – 0.7 for a density of 1600 – 1800 kg/m^3 [5, 21].

The process of formation of a continuous hollow fiber is based on applying compressed air to the fluid glass bulb, using a nozzle located inside the spinneret, concentric to the nozzle opening. Compressed air forms an air cavity at the end of the spinneret, around which the glass melt drawn from the annular slot cools, thus forming a hollow fiber.

Hollow fibers have a higher bending hardness than solid fibers. The hardness depends on the capillary coefficient and the fiber diameter.

Testing of composites based on hollow fibers revealed the advantages of the new type of fibers in thermophysical and dielectric properties, which determined its efficient application in heat-insulating and radio-transmitting materials. The bending hardness of laminar fiberglass plastics reinforced with hollow fiber can be doubled, as compared with the hardness of standard fiberglass plastics. Owing to the lower density of plastics based on hollow fiber, their thermal conductivity is two times lower, and their temperature conductivity and dielectric permittivity are lower by 35% [21].

By modifying the chemical composition of glass for hollow fiber, it is possible to obtain materials whose dielectric permittivity approaches similar values in quartz glass. These

materials are especially promising for the production of radar housing.

Quartz glass has a set of unique physicochemical properties. It surpasses other types of continuous glass fibers in strength, chemical resistance, dielectric parameters, and temperature resistance. These properties are due to the chemical composition of quartz glass: the SiO_2 content in quartz fiber is about 99.95 wt.%. Since the Si–O bond has high energy, the strength of quartz fibers is higher than the strength of high-strength type-S fiber. The softening point of such fiber is equal to 1667°C; therefore, materials made of this fiber can serve for a long time at temperatures up to 1200°C and withstand short-term impacts up to 2000°C [5, 21].

The quartz fiber technology fundamentally differs from other fiber technologies. It is virtually impossible to draw quartz fiber through a spinneret. This fiber is produced by drawing quartz molding fillets. The expensive technology of producing quartz beads (fillets) and their drawing determines the high cost of this type of fiber. However, the unique properties of quartz glass are the reason for its wide application in the nuclear and space industries and in radioelectronics, chemistry, and optics.

Thus, the use of glass fiber in various fields of industry and economics and various requirements imposed on fiber determine the wide range of glass systems and compositions. Despite of considerable progress in this field, studies directed toward the improvement and modification of fiberglass compositions are being continued in numerous research and production centers in different countries. The main challenges confronting researchers are as follows: development of glass fibers with a set of high service parameters; improvement of the technological properties of the compositions; solving environmental problems involved in production; decreasing the production cost of fiber and fiber articles. The contemporary level of glass science and glass-melting technology allows one to hope for the successful solution of these important problems.

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